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# Ultrasonic studies in binary liquid mixtures

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**Abstract** Densities and ultrasonic speeds of pure acetonitrile (ACN), 1-butanol, formamide (FA) and those of eighteen binary mixtures of ACN with 1-butanol and FA were measured at 308 15 K From the experimental data, isentropic compressibility, intermolecular free length, relative association, acoustic impedance, molar sound speed, excess isentropic compressibility, excess intermolecular free length, excess volume, deviation in ultrasonic speed and excess acoustic impedance were calculated The variation of these parameters with composition of the mixtures suggest that for the system ACN + 1-butanol structure-breaking effect dominates over that of the hydrogen-bonding effect between unlike molecules, while the system ACN + FA shows opposite behaviour with composition Furthermore, theoretical values of ultrasonic speed in these systems were calculated using different theories and relations Their suitability for the present systems were examined

**Keywords** Binary liquid mixtures, acoustical parameters, molecular interactions

**PACS Nos.** 43 35 Bf, 62 60 +v

## 1. Introduction

The practical applications of mixed solvents, rather than single solvent, in industrial and biological processes have been recognised all over the world, as they provide a wide choice of solutions with appropriate properties. Ultrasonics has proved to be a useful tool for gaining information on the dynamics of these liquid systems. The measurement of ultrasonic speed enables the accurate determination of some useful parameters and their excess functions, which are highly sensitive to molecular interactions, and can be used to provide qualitative information about the physical nature and strength of molecular interaction in these liquid mixtures [1–3]. In continuation of our earlier studies [4–7] on intermolecular interactions in non-aqueous binary liquid mixtures containing protic and aprotic solvents, here we report the results of our study on the binary mixtures of acetonitrile (ACN) with 1-butanol and formamide (FA), covering the entire composition range, at 308.15 K. It is well known that acetonitrile, 1-butanol and formamide are associated in pure state. ACN molecules are highly polar (dipole moment $\mu$ = 3.92 D at 293.15 K) [8], non-hydrogen bonded, with parallel and anti-parallel orientations and this strongly ordered structure is stabilized by dipole-dipole interactions [9,10]. 1-Butanol molecules are also polar ($\mu$ = 1.66 D at 293.15 K) [8] and self-associated through hydrogen bonding of their hydroxyl groups [11], whereas FA molecules are highly polar ($\mu$ = 3.73 D at 293.15 K) [8] and are strongly self-associated through extensive network of hydrogen bonds [12,13]. The applications of these solvents and their mixtures in chemistry and modern technology [14] and close relationship of FA to peptide and proteins led us to undertake the present investigation.

In the present investigation, we report densities, $\rho$ and ultrasonic speeds, $u$ of the mixtures of ACN with 1-butanol and FA, covering the entire composition range, at 308.15 K. From these experimental data, isentropic compressibilities $K_s$, intermolecular free lengths $L_f$, relative associations $R_A$, acoustic impedances $Z$, molar sound speeds $R_m$, excess isentropic compressibilities $K_s^E$, excess intermolecular free lengths $L_f^E$, excess volumes $V^E$, deviations in ultrasonic speeds $\Delta u$ and excess acoustic impedances $Z^E$ were calculated. The dependence of these parameters on composition of the mixture reveals the nature and extent of interaction between the component molecules, which enables us to have a

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better understanding of solution behaviour of the liquid mixtures.

Ultrasonic speeds were theoretically calculated by using free length theory (FLT) [15] collision factor theory (CFT) [16], Nomoto's relation [17] and Van Dael and Vangeel's relation [18]. The relative merits of these theories and relations were examined by comparing the theoretical values of ultrasonic speeds with the values obtained experimentally.

## 2. Experimental

ACN (s.d fine, India) was the same as used in our previous study [19]. 1-Butanol and FA both (s.d. fine, India) of AR and spectroscopic grade, respectively, were purified by standard procedures described in the literature [20, 21].

The mixtures were prepared by weight and were kept in air-tight bottles. The weighings were done on Afcoset ER-120A electronic balance with a precision of ±0.1 mg. The probable error in mole fraction was estimated to be less than ±0.0001. The densities of the pure liquids and their binary mixtures were measured using a single-capillary pycnometer (made of Borosil glass). The marks on the capillary were calibrated by using triple distilled water. The accuracy in density measurements was found to be ±0.0001 g cm $^{3}$. The ultrasonic speeds were measured using single-crystal variable-path ultrasonic interferometer (Mittal Enterprise, New Delhi, M-82) at 3 MHz with an accuracy of ± 0.05%. The temperature of the samples was maintained at 308.15 ± 0.05 K in an electronically controlled thermostatic water bath.

## 3. Results and discussion

The experimental values of density, $\rho$ and ultrasonic speed, $u$ of pure liquids and their binary mixtures have been used to calculate the values of isentropic compressibility $K_s$, intermolecular free length $L_f$, relative association $R_A$, acoustic impedance $Z$ and molar sound speed $R_m$ using the following standard relations :

$$K_s = 1/u^2 \rho, \tag{1}$$

$$L_f = K/u\rho^{1/2}, \tag{2}$$

$$R_A = (\rho/\rho_0)(u_0/u)^{1/3}, \tag{3}$$

$$Z = u\rho, \tag{4}$$

$$R_m = Vu^{1/3}, \tag{5}$$

where $K$ is a temperature dependent constant [= (93.875 + 0.375 $T$) × $10^{-8}$], $T$ is the absolute temperature $\rho_0$, $\rho$ and $u_0$, $u$ are the densities and ultrasonic speeds of the solvent (1-butanol/FA) and solution, respectively; $V$ is the molar volume of the binary mixtures, calculated using the equation

$$V = [(1-x)M_1 + xM_2]/\rho, \tag{6}$$

where $M_1$ and $M_2$ are the molecular weights of 1-butanol/FA and ACN, respectively. The values of $\rho$, $u$, $K_s$, $L_f$, $R_A$, $Z$ and $R_m$ as a function of mole fraction $x$ of ACN at 308.15 K are listed in Table 1. The non-linear increase/decrease in $u$, $K_s$, $L_f$, $R_A$, $Z$ and $R_m$ values (Table 1) with composition for both the systems (ACN + 1-butanol/FA) indicates significant interactions between the component molecules [22, 23]. The observed increase in $u$ and corresponding decrease in $L_f$ for ACN + 1-butanol, while a decrease in $u$ (with an increase in $L_f$) for ACN + FA as a result of mixing of component liquids are in accordance with the view proposed by Eyring and Kincaid [24], according to which the ultrasonic speed increases with decrease in $L_f$ and *vice-versa*.

The excess functions are found to be more sensitive towards intermolecular interactions in liquid mixtures. The excess isentropic compressibility $K_s^E$, excess intermolecular free length $L_f^E$, excess volume $V^E$, deviations in ultrasonic speed $\Delta u$ and excess acoustic impedance $Z^E$ have been calculated using the following relation :

$$Y^E = Y - [(1-x)Y_1 + xY_2], \tag{7}$$

where $Y^E$ is $K_s^E$ or $L_f^E$ or $V^E$ or $\Delta u$ or $Z^E$; $x$ is the mole fraction of ACN (volume fraction, $\phi$ is used in case of $K_s^E$) and subscripts 1 and 2 stand for 1-butanol/FA and ACN, respectively. These excess functions were fitted to a Redlich-Kister [25] type polynomial equation

$$Y^E = x(1-x)\sum_{i=1}^{5} A_i(1-2x)^i. \tag{8}$$

The values of coefficients $A_i$ evaluated by the method of least-squares are given in Table 2 along with the standard deviations $\sigma(Y^E)$. The variation in $K_s^E$, $L_f^E$, $V^E$, $\Delta u$ and $Z^E$ with mole fraction $x$ of ACN are presented graphically in Figures 1–5, respectively.

Figures 1, 2 and 3 show that $K_s^E$, $L_f^E$ and $V^E$ are positive for ACN + 1-butanol, while negative for ACN + FA binary mixtures over the whole composition range. Positive or negative deviations in these functions from rectilinear dependence on composition for the present systems indicate the extent of interaction between unlike molecules. The observed values of $K_s^E$, $L_f^E$ and $V^E$ can be qualitatively explained by considering the factors which influence these excess functions, namely, (i) the mutual disruption of associates present in the pure liquids (dipolar association of ACN and

**Table 1.** Values of density ($\rho$), ultrasonic velocity ($u$), isentropic compressibility ($K_s$), intermolecular free length ($L_f$), relative association ($R_A$), acoustic impedance ($Z$) and molar sound speed ($R_m$) of binary mixtures of ACN with 1-butanol and FA at 308.15 K.

| $x$ (ACN) | $\rho$ (kg m$^{-3}$) | $u$ (m s$^{-1}$) | $K_s$ ($10^{-10}$ m$^2$ N$^{-1}$) | $L_f$ ($10^{-11}$ m) | $R_A$ | $Z$ ($10^6$ kg m$^{-2}$ s$^{-1}$) | $R_m$ [$10^{-4}$ m$^3$ mol$^{-1}$ (m s$^{-1}$)$^{1/3}$] |
|---|---|---|---|---|---|---|---|
| | | | ACN + 1-Butanol | | | | |
| 0 0000 | 798 4 | 1205.2 | 8 6231 | 6 1500 | 1.0000 | 0 9622 | 9.8795 |
| 0 1615 | 794.5 | 1207.5 | 8.6324 | 6 1533 | 0.9945 | 0 9594 | 9.2185 |
| 0 3023 | 790 6 | 1209.6 | 8.6449 | 6 1577 | 0 9890 | 0.9563 | 8 6416 |
| 0 4262 | 786 8 | 1212.0 | 8 6523 | 6 1604 | 0.9836 | 0 9536 | 8.1338 |
| 0 5361 | 783 2 | 1215.1 | 8.6478 | 6 1588 | 0 9783 | 0 9517 | 7.6832 |
| 0 6342 | 779 8 | 1218.6 | 8.6356 | 6.1544 | 0 9731 | 0 9503 | 7.2799 |
| 0 7222 | 776 6 | 1222.3 | 8.6188 | 6 1484 | 0.9681 | 0 9492 | 6 9163 |
| 0 8018 | 773 7 | 1226 7 | 8 5892 | 6.1379 | 0 9634 | 0.9491 | 6 5866 |
| 0 8740 | 771 1 | 1232 2 | 8 5414 | 6 1208 | 0 9587 | 0 9501 | 6 2868 |
| 0 9398 | 768 6 | 1238 3 | 8 4849 | 6 1005 | 0 9540 | 0 9518 | 6 0136 |
| 1 0000 | 766 5 | 1245 1 | 8 4155 | 6 0755 | 0 9497 | 0.9544 | 5.7615 |
| | | | ACN + FA | | | | |
| 0 0000 | 1121 1 | 1577.2 | 3 5858 | 3 9658 | 1.0000 | 1 7682 | 4 6764 |
| 0.0770 | 1088 3 | 1545 5 | 3 8469 | 4 1077 | 0 9773 | 1 6820 | 4 7523 |
| 0 1580 | 1056 1 | 1512 4 | 4 1396 | 4 2611 | 0.9553 | 1.5972 | 4.8268 |
| 0 2433 | 1022 9 | 1477 2 | 4 4801 | 4 4329 | 0 9325 | 1.5110 | 4.9066 |
| 0 3334 | 988 7 | 1443 4 | 4 8547 | 4 6145 | 0 9084 | 1.4271 | 4.9962 |
| 0 4287 | 953 7 | 1409 1 | 5 2809 | 4.8128 | 0 8832 | 1.3439 | 5.0935 |
| 0 5295 | 917 4 | 1375 4 | 5.7621 | 5 0273 | 0.8565 | 1 2618 | 5.2038 |
| 0 6365 | 880 1 | 1340 4 | 6.3241 | 5 2667 | 0 8288 | 1.1797 | 5.3244 |
| 0 7501 | 842.5 | 1305 7 | 6 9621 | 5 5260 | 0.8003 | 1 1001 | 5.4548 |
| 0 8710 | 804 3 | 1274 5 | 7 6542 | 5.7942 | 0.7702 | 1 0251 | 5.6030 |
| 1 0000 | 766 5 | 1245 1 | 8 4155 | 6 0755 | 0 7398 | 0 9544 | 5 7615 |

**Table 2.** Coefficients $A_i$ of eq. (7) along with standard deviations $\sigma(Y^E)$ of binary mixtures

| Properties | $A_1$ | $A_2$ | $A_3$ | $A_4$ | $A_5$ | $\sigma(Y^E)$ |
|---|---|---|---|---|---|---|
| | | | ACN + 1-Butanol | | | |
| $K_s^E$ ($10^{-10}$ m$^2$ N$^{-1}$) | 0.4638 | –0 1170 | 0.2035 | –0 0182 | –0 3795 | 0 0004 |
| $L_f^E$ ($10^{-12}$ m) | 1 8603 | –1 2064 | 0.9166 | –0 5913 | –0.7270 | 0 0068 |
| $V^E$ ($10^{-6}$ m$^3$ mol$^{-1}$) | 0 8729 | –0.5115 | –0.1034 | 0 0657 | 0 3281 | 0.0018 |
| $\Delta u$ (m s$^{-1}$) | –44.248 | 22.488 | –19 213 | 12 348 | 16.847 | 0 1206 |
| $Z^F$ ($10^5$ kg m$^{-2}$ s$^{-1}$) | –0 2395 | 0.2087 | –0.1312 | 0 0856 | 0.0712 | 0 0012 |
| | | | ACN + FA | | | |
| $K_s^E$ ($10^{-10}$ m$^2$ N$^{-1}$) | –2.8846 | 0.5616 | 0.1847 | –0.2683 | –0.0350 | 0 0008 |
| $L_f^E$ ($10^{-12}$ m) | –2.2334 | –1 1395 | 1.4286 | –0 2911 | –1 0967 | 0.0072 |
| $V^E$ ($10^{-6}$ m$^3$ mol$^{-1}$) | –1 8939 | –0 4440 | 0 6230 | 0 4596 | 0.2176 | 0.0018 |
| $\Delta u$ (m s$^{-1}$) | –105.36 | 10 467 | –27.065 | 12.241 | 44.099 | 0.2876 |
| $Z^E$ ($10^5$ kg m$^{-2}$ s$^{-1}$) | –3 0543 | –0.1172 | –0.3423 | –0 0663 | 0 2605 | 0.0024 |

hydrogen bonds in 1-butanol/FA), (ii) the formation of new hydrogen bonds between unlike molecules (between nitrogen of ACN, with its lone pair of electrons, and hydrogen atoms of –OH and $–NH_2$ groups of 1-butanol and FA, respectively, and (iii) geometrical fitting of component molecules into each others structure. The first factor contributes to positive

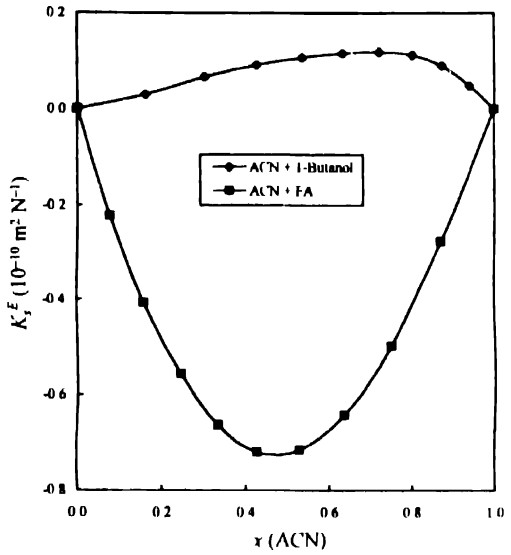


**Figure 1.** Variation of excess isentropic compressibility $K_s^E$ with mole fraction of acetonitrile (ACN) for the binary mixtures at 308 15 K

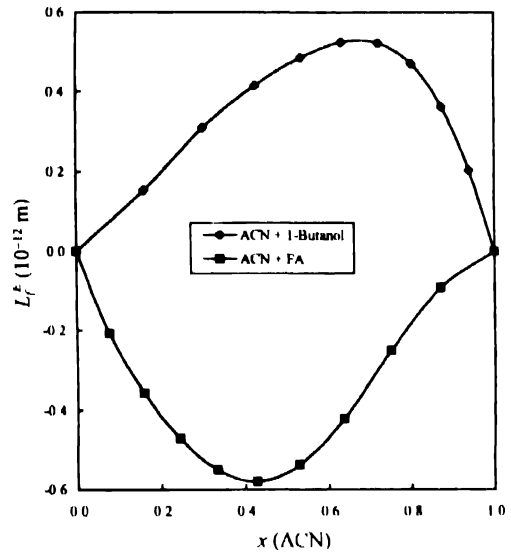


**Figure 2.** Variation of excess intermolecular free length $L_f^E$ with mole fraction of acetonitrile (ACN) for the binary mixtures at 308 15 K

$K_s^E$, $L_f^E$ and $V^E$, while the remaining two factors lead to negative $K_s^E$, $L_f^E$ and $V^E$ values. The observed positive values of $K_s^E$, $L_f^E$ and $V^E$ (Figures 1–3), over whole composition range, for ACN + 1-butanol binary mixtures suggest that the effect due to break up of dipolar associations of ACN and hydrogen bonds of 1-butanol predominates over that of hydrogen bonding between unlike molecules, making $K_s^E$, $L_f^E$ and $V^E$ values positive. The positive $K_s^E$, $L_f^E$ and $V^E$ values may also be attributed to the weak dipole-dipole interaction leading to weak complex formation [26] between unlike molecules. It is interesting to note that $K_s^E$, $L_f^E$ and $V^E$ values are more positive in ACN rich region. This shows that the mutual dissociation of ACN and 1-butanol associates is more pronounced in ACN-rich region. Though there is a

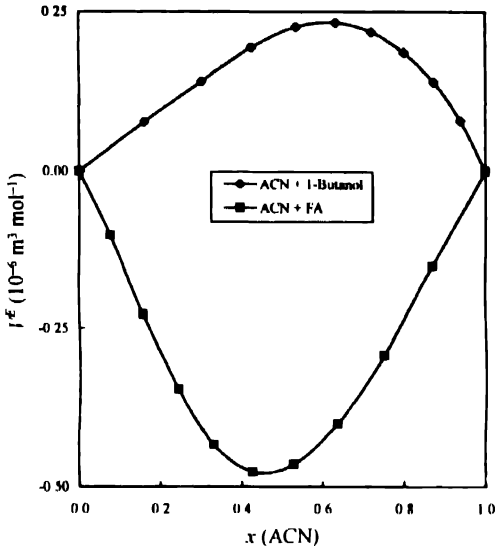


**Figure 3.** Variation of excess volume $V^E$ with mole fraction of acetonitrile (ACN) for the binary mixtures at 308 15 K

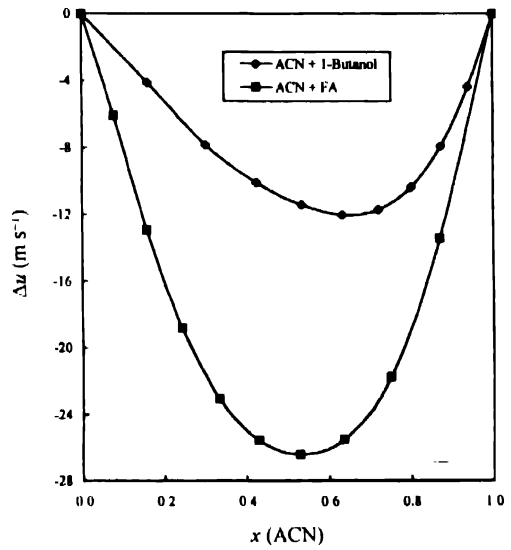


**Figure 4.** Variation of deviation in sound speed $\Delta u$ with mole fraction of acetonitrile (ACN) for the binary mixtures at 308 15 K

considerable difference between the molar volumes of ACN and 1-butanol, the contribution due to fitting of smaller ACN molecules into the structure of bigger 1-butanol molecules and the possibility of negative $K_s^E$, $L_f^E$ and $V^E$ values seems to be insignificant. Similar results have also been reported by others for methanol + 1-butanol [27], toluene + 1-butanol [28] and ACN + bromoform [29] binary mixtures.

The negative $K_s^E$, $L_f^E$ and $V^E$ values (Figures 1–3) for ACN + FA mixtures indicate that the formation of H-bonds (between nitrogen atom of ACN and hydrogen atoms of H–N–H group of FA) dominates over the disruption of associates present in the pure liquids. Another source of negative contribution to $K_s^E$, $L_f^E$ and $V^E$ may be from the fitting of ACN molecules into the voids created by three dimensional hydrogen bonded networks of FA molecules. Contributions arising from the fitting of smaller alkanol molecules into the voids available in the structure of FA were also considered [23,30] for interpreting the negative $V^E$ values of FA + alkanols binary mixtures.

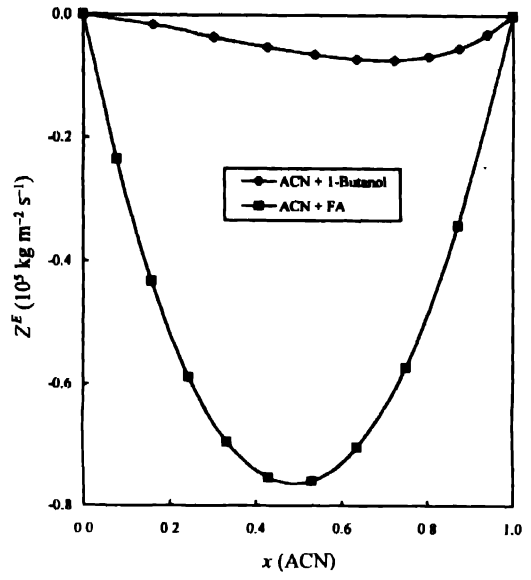


**Figure 5.** Variation of excess acoustic impedance $Z^E$ with mole fraction of acetonitrile (ACN) for the binary mixtures at 308.15 K.

As expected, Figures 4 and 5 show that the behaviours of $\Delta u$ and $Z^E$ support each other. Both exhibit negative deviations over the entire composition range for the systems under study. Negative deviations in $\Delta u$ and $Z^E$ from rectilinear dependence on composition suggest that dispersion forces are also operative in these systems. This seems to be in good agreement with the conclusion arrived at by Fort and Moore [22] for the study of excess functions for binary liquid mixtures. Negative deviations in $\Delta u$ and $Z^E$ have also been reported for dimethylsulphoxide + alkanols [31]/toluene [32] binary liquid mixtures.

**Table 3.** Theoretical values of ultrasonic speed calculated from CFT, FLT, Nomoto's and Van Dael and Vangeel's equation along with the experimental values of ultrasonic speed and percentage error for the binary mixtures at 308 15 K.

| $x$ (ACN) | $u_{expt}$ (m s$^{-1}$) | $u_{FLT}$ (m s$^{-1}$) | $u_{CFT}$ (m s$^{-1}$) | $u_{NOM}$ (m s$^{-1}$) | $u_{VD}$ (m s$^{-1}$) | % error FLT | CFT | NOM | VD |
|---|---|---|---|---|---|---|---|---|---|
| | | | | ACN + 1-Butanol | | | | | |
| 0 0000 | 1205.2 | 1205 2 | 1205 2 | 1205 2 | 1205.2 | 0 00 | 0.00 | 0 00 | 0 00 |
| 0 1615 | 1207 5 | 1205 1 | 1210 6 | 1209 2 | 1186 6 | 0 20 | 0 26 | 0 14 | 1 73 |
| 0 3023 | 1209 6 | 1204.9 | 1215 2 | 1213 1 | 1178 4 | 0.39 | 0 46 | 0.29 | 2 58 |
| 0 4262 | 1212 0 | 1205.2 | 1219.2 | 1217 1 | 1176.9 | 0 56 | 0.59 | 0.42 | 2.89 |
| 0 5361 | 1215 1 | 1206 6 | 1222 9 | 1221.1 | 1180.1 | 0.70 | 0.64 | 0.49 | 2.88 |
| 0 6342 | 1218 6 | 1209.0 | 1226 4 | 1225 0 | 1186 6 | 0 79 | 0 64 | 0 53 | 2 63 |
| 0 7222 | 1222 3 | 1212.7 | 1229.9 | 1229 0 | 1195 5 | 0 79 | 0.62 | 0 55 | 2.20 |
| 0 8018 | 1226 7 | 1218 2 | 1233.5 | 1233.0 | 1206 1 | 0.70 | 0 55 | 0.52 | 1.68 |
| 0 8740 | 1232.2 | 1225.6 | 1237.3 | 1237 1 | 1218 1 | 0 53 | 0 41 | 0.39 | 1.14 |
| 0 9398 | 1238 3 | 1233 9 | 1241.0 | 1241.1 | 1231 2 | 0 35 | 0 22 | 0 22 | 0.57 |
| 1 0000 | 1245.1 | 1245.1 | 1245.1 | 1245.1 | 1245 1 | 0.00 | 0.00 | 0.00 | 0 00 |
| | | | | ACN + FA | | | | | |
| 0 0000 | 1577.2 | 1577 2 | 1577 2 | 1577 2 | 1577.2 | 0 00 | 0.00 | 0.00 | 0 00 |
| 0 0770 | 1545 5 | 1343 2 | 1555 4 | 1541 6 | 1538.2 | 13.09 | 0.64 | 0.25 | 0 47 |
| 0.1580 | 1512.4 | 1308 2 | 1533.3 | 1506.5 | 1500.7 | 13.50 | 1 38 | 0.39 | 0 77 |
| 0.2433 | 1477.2 | 1287.2 | 1508 4 | 1472.0 | 1464 7 | 12 86 | 2.11 | 0 35 | 0 85 |
| 0 3334 | 1443 4 | 1272.6 | 1480.5 | 1438.1 | 1430 0 | 11 84 | 2.57 | 0 37 | 0 93 |
| 0 4287 | 1409.1 | 1263 5 | 1449 8 | 1404 6 | 1396.4 | 10.33 | 2 89 | 0.32 | 0.90 |
| 0 5295 | 1375.4 | 1252 2 | 1415 2 | 1371 7 | 1364 1 | 8 96 | 2 90 | 0 27 | 0.82 |
| 0.6365 | 1340 4 | 1242 7 | 1377 1 | 1339.2 | 1332 8 | 7 29 | 2.74 | 0 09 | 0 57 |
| 0 7501 | 1305.7 | 1239.2 | 1336.1 | 1307.4 | 1302.6 | 5.09 | 2 33 | 0 13 | 0 24 |
| 0 8710 | 1274.5 | 1238.0 | 1291 7 | 1276.0 | 1273 4 | 2.86 | 1 35 | 0 12 | 0 09 |
| 1 0000 | 1245.1 | 1245.1 | 1245 1 | 1245 1 | 1245.1 | 0.00 | 0 00 | 0 00 | 0.00 |

The theoretical values of ultrasonic speeds in the mixtures : ACN + 1-butanol and + FA were calculated using the following theories and relations :

Free length theory [15] :

$$u_{FLT} = K/(L_f \rho^{1/2}), \tag{9}$$

Collision factor theory [16] :

$$u_{CFT} = u_\infty \left(x_1 S_1 + x_2 S_2\right)\left[\left(x_1 B_1 + x_2 B_2\right)/V\right], \tag{10}$$

Nomoto's relation [17] :

$$u_{NOM} = \left[\left(x_1 R_1 + x_2 R_2\right)/\left(x_1 V_1 + x_2 V_2\right)\right]^3, \tag{11}$$

Van Dael and Vangeel relation [18] :

$$u_{VD} = \left\{\left[\left(x_1/M_1 u_1^2\right) + \left(x_2/M_2 u_2^2\right)\right] \times \left(x_1 M_1 + x_2 M_2\right)\right\}^{-1/2} \tag{12}$$

The details of derivation and terms used may be obtained from the literature [15-18,33,34]. The theoretical values of ultrasonic speeds along with the experimental values and percentage errors in the calculated values are summarised in Table 3. On comparison, the results indicate that for ACN + 1-butanol mixtures Nomoto's relation, CFT and FLT provide good agreement between theoretical and experimental ultrasonic speeds, while Van Dael and Vangeel relation shows maximum deviation. For ACN + FA binary mixtures Nomoto's relation seems to provide best results followed by Van Deal and Vangeel relation, CFT and FLT.

It may be concluded that out of the four theories and relations discusses above the Nomoto's relation provides the best results for ultrasonic behaviour of the present systems. Thus, the linearity of molecular sound speed and additivity of molar volumes, as suggested by Nomoto [17] in deriving the empirical relation (11), have been truely observed in the aforementioned binary liquid mixtures.

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